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Elastic Anomalies in Orbital-Degenerate Frustrated Spinel CoV_2O_4

Tadataka Watanabe[1*], Shogo Yamada[1], Rui Koborinai[2], Takuro Katsufuji[2]
[1] *Department of Physics, College of Science and Technology, Nihon University, Chiyoda, Tokyo 101-8308, Japan*
[2] *Department of Physics, Waseda University, Shinjuku, Tokyo 169-8555, Japan*

Abstract
Ultrasound velocity measurements were performed on a single crystal of the orbital-degenerate frustrated spinel CoV_2O_4 in all the symmetrically-independent elastic moduli of the cubic crystal. The measurements of temperature dependence of the elastic moduli observed discontinuous elastic anomalies due to a ferrimagnetic transition at T_C = 165 K and another phase transition at T^* = 50 K. Additionally, the measurements observed anomalous temperature dependence of the elastic moduli, specifically, non-monotonic temperature dependence in the magnetically-ordered phase below T_C, and magnetic-field-sensitive elastic softening with decreasing temperature in the paramagnetic phase above T_C. These anomalous temperature variations below and above T_C should be driven by the coupling of lattice to magnetic excitations.

Keywords: Vanadate spinel, Frustration, Orbital order, Sound velocity

1 Introduction

Vanadate spinels AV_2O_4 (A = Zn [1], Mg [2], Cd [3], Mn [4], Fe [5], and Co [6]) are geometrically frustrated magnets in which the pyrhochlore lattice consists of V^{3+} ($3d^2$) ions with t_{2g} orbital degeneracy. AV_2O_4 undergoes a magnetic phase transition, specifically, an antiferromagnetic transition for nonmagnetic A = Zn (T_N = 40 K [1]), Mg (T_N = 42 K [2]), and Cd (T_N = 33 K [3]), and a ferrimagnetic transition for magnetic A = Mn (T_C = 57 K [4]), Fe (T_C = 105 K [5]), and Co (T_C ~ 150 K [6]). For AV_2O_4, it is considered that the orbital degrees of freedom play an important role for the occurrence of the magnetic transition.

CoV_2O_4 is the only vanadate spinel which exhibits the absence of structural phase transition down to low temperature, while this compound exhibits a ferrimagnetic phase transition at T_C ~ 150 K [6]. And it is also noted that CoV_2O_4 exhibits the highest electrical conductivity among AV_2O_4 [6]. The

* Corresponding author. Tel: +81-3-3259-0920; fax: +81-3-3293-8269
E-mail address: tadataka@phys.cst.nihon-u.ac.jp

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absence of structural phase transition and the nearly-metallic character in CoV_2O_4 are both expected to be results of the interplay between frustration and orbital degrees of freedom.

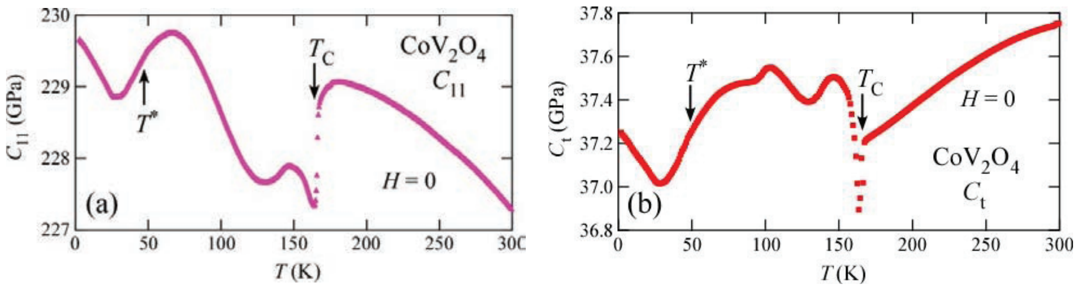
We performed ultrasound velocity measurements in CoV_2O_4 to study the interplay of orbital, spin, and lattice degrees of freedom. The sound velocity or the elastic modulus is a useful probe enabling symmetry-resolved thermodynamic information to be extracted from frustrated magnets [7,8,9,10,11]. The present study in CoV_2O_4 observed a variety of elastic anomalies which should have magnetic origins.

2 Experimental

The ultrasound velocity measurements were performed on a single crystal of CoV_2O_4 grown by the floating-zone method. Temperature (T) dependence of the ultrasound velocity was measured at T from 2 K to 300 K with magnetic field H||[110] up to 7 T in all the symmetrically-independent elastic moduli of the cubic crystal: compression modulus C_{11}, tetragonal shear modulus $(C_{11}\text{-}C_{12})/2 = C_t$, and trigonal shear modulus C_{44}. The respective measurements of C_{11}, C_t, and C_{44} were performed using longitudinal sound waves with propagation $\boldsymbol{k}$||[001] and polarization $\boldsymbol{u}$||[001], transverse sound waves with $\boldsymbol{k}$||[110] and $\boldsymbol{u}$||[1-10], and transverse sound waves with $\boldsymbol{k}$||[110] and $\boldsymbol{u}$|| [001]. The sound velocities of CoV_2O_4 measured at room temperature (300 K) are 6740 m/s for C_{11}, 2750 m/s for C_t, and 3770 m/s for C_{44}.

3 Results and discussion

Figures 1(a), (b) and (c) respectively show T dependence of compression modulus $C_{11}(T)$ tetragonal shear modulus C_t (T), and trigonal shear modulus $C_{44}(T)$ in CoV_2O_4 without magnetic field (H = 0). All the elastic modes exhibit a discontinuous change at T_C = 165 K as marked by arrows in Figs. 1(a)-(c), which is due to the ferrimagnetic transition. Additionally, only $C_{44}(T)$ exhibits a discontinuous change at T^* = 50 K as marked by arrow in Fig. 1(c), indicating the occurrence of another phase transition at T^* which sensitively couples to the trigonal lattice deformations. This



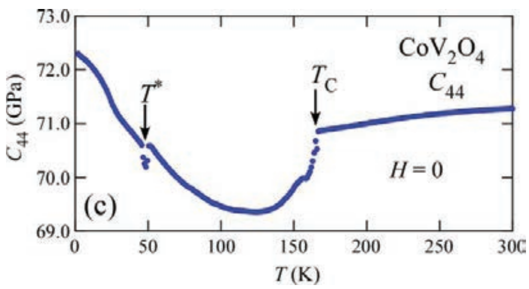
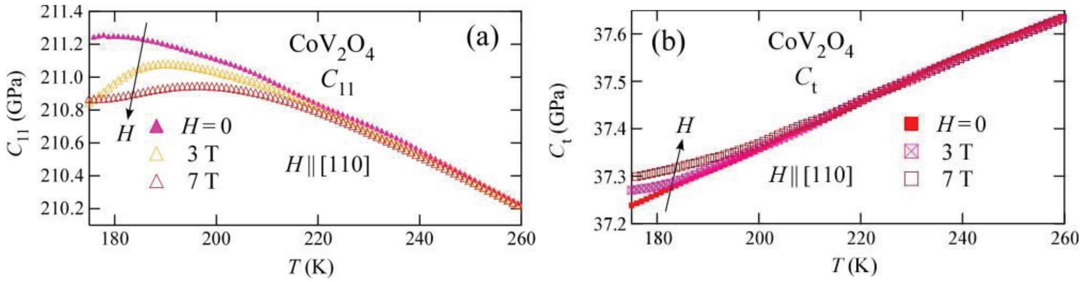


Figure 1: Temperature dependence of elastic moduli in CoV_2O_4 without magnetic field (H = 0). (a) Compression modulus C_{11}, (b) tetragonal shear modulus $(C_{11}\text{-}C_{12})/2 = C_t$, and (c) trigonal shear modulus C_{44}. T_C and T^* are marked by arrows in (a)-(c).

transition at T^* should correspond to the disorder-sensitive phase transition which was observed in the specific heat measurements [6,12].

As shown in Figs. 1(a)-(c), below T_C, all the elastic modes exhibit non-monotonic T dependence with characteristic minima at ~30 K and ~130 K for $C_{11}(T)$, ~30 K, ~100 K, and ~130 K for $C_t(T)$, and ~130 K for $C_{44}(T)$. Such an elastic-modulus-minimum anomaly in $C_\Gamma(T)$ usually arises from the coupling of lattice to gapped magnetic excitations [8,9,11]. Although the direct observation of magnetic excitations in CoV_2O_4 has not been reported yet, the elastic-modulus-minimum anomalies in $C_\Gamma(T)$ of CoV_2O_4 should stem from the coupling of lattice to gapped magnetic excitations. Since the temperature at which $C_\Gamma(T)$ minimum occurs is considered to correspond roughly to the gap magnitude of the excitations, the presence of the multiple $C_\Gamma(T)$ minima in the magnetically-ordered phase of CoV_2O_4 suggests the coupling of lattice to multiple gapped excitation modes.

In the paramagnetic phase ($T > T_C$), as shown in Figs. 1(a)-(c), $C_t(T)$ and $C_{44}(T)$ exhibit softening with decreasing T while $C_{11}(T)$ exhibits hardening with decreasing T. Figures 2(a), (b), and (c) show $C_{11}(T)$, $C_t(T)$, and $C_{44}(T)$ with $H\|[110]$ in the paramagnetic phase ($T > T_C$), respectively. It is evident that $C_\Gamma(T)$ varies with H in all the elastic modes. We note here that MnV_2O_4 and MgV_2O_4 exhibit H-sensitive elastic softening with decreasing T in the paramagnetic phase [10,11]. While $C_\Gamma(T)$ of MnV_2O_4 exhibits huge ~-1/T-type softening (convex T dependence) in the paramagnetic phase which is considered to be driven by orbital-spin-coupled fluctuations [10], $C_\Gamma(T)$ of MgV_2O_4 exhibits concave T dependence which is considered to be driven by orbital-spin-coupled gapped excitations [11]. Since the H-sensitive elastic softening of $C_t(T)$ and $C_{44}(T)$ in the paramagnetic phase of CoV_2O_4 resembles that of MgV_2O_4, this elastic softening in CoV_2O_4 should be attributed to orbital-spin-coupled gapped excitations. The H-sensitivity of $C_{11}(T)$ shown in Fig. 2(a) indicates that this elastic mode also contains the component of the H-sensitive softening which should be driven by the coupling of lattice to orbital-spin excitations, although the component of the H-sensitive softening in $C_{11}(T)$ is weaker than that of ordinal hardening (background) in this elastic mode. It is noted here that, as indicated by arrows in Figs. 2(a)-(c), while the softening in $C_t(T)$ and $C_{44}(T)$ is relaxed with increasing H, the softening component in $C_{11}(T)$ is enhanced with increasing H. Thus it is suggested that the softening component in $C_{11}(T)$ originates from orbital-spin-coupled excitations which are different from the origin of the softening in $C_t(T)$ and $C_{44}(T)$. Taking into account the absence of structural phase transition in CoV_2O_4 in contrast to its presence in MnV_2O_4 and MgV_2O_4, it is



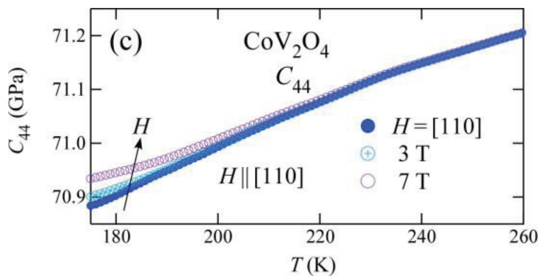


Figure 2: Temperature dependence of elastic moduli in the paramagnetic phase of CoV_2O_4 ($T > T_C$) with $H\|[110]$. (a) $C_{11}(T)$, (b) $C_t(T)$, and (c) $C_{44}(T)$. The arrows in (a)-(c) are guides to the eye, indicating the variations of $C_{11}(T)$, $C_t(T)$, and $C_{44}(T)$ with increasing H.

expected that the orbital-spin-coupled excitations in CoV_2O_4 is inherently different from those in MnV_2O_4 and MgV_2O_4.

4 Summary

In summary, ultrasound velocity measurements in the orbital-degenerate frustrated spinel CoV_2O_4 observed a variety of elastic anomalies which should have magnetic origins. Discontinuous anomalies in T dependence of the elastic moduli at T_C = 165 K and T^* = 50 K should be due to a ferrimagnetic transition and another phase transition, respectively. Non-monotonic T dependence of the elastic moduli in the magnetically-ordered phase below T_C should be due to the coupling of lattice to gapped magnetic excitations. And H-sensitive elastic softening with decreasing T in the paramagnetic phase above T_C should be due to the coupling of lattice to orbital-spin-coupled excitations. Further experimental and theoretical studies are indispensable to understand the phase transitions and the magnetic excitations in CoV_2O_4.

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